

Manipulation of form birefringence in isotropic material

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Abstract: Form-birefringent nanostructure composed of the self-organized oxygen defects can be created by light pulses with a width of 70 fs. Such rewritable and directionally controllable nanostructures have evolved by lowering threshold for defect formation.

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Material processing with ultrafast lasers has recently attracted considerable interest [1] due to a wide range of applications including laser surgery [2], 3D micro- and nano-structuring [3]. Here we experimentally demonstrate the self-organized oxygen defects in fused silica are rewritable and directionally controllable with femtosecond pulses. Such self-organization takes place when the photo-induced stress exceeds a certain threshold and develops initially from residual birefringence originated from internal stress due to thermal distribution with steep gradients. The stress accumulation and the oxygen defect generation depend on the polarization direction, which could be explained by the anisotropy of electron plasma absorption for p and s polarizations at the oblique, moving with the speed of light interface produced by the pulse with tilted intensity front. Such anisotropy affects the reciprocal evolution of the material modification in isotropic medium, finally exhibits a non-reciprocal *quill writing effect* [4] revealing strong dependence of the material modification on the orientation of writing direction relative to the direction of pulse front tilt.

The experiments were performed using a mode-locked, regeneratively amplified Ti: Sapphire laser system (Coherent RegA 9000), operating at 800 nm with 70 fs pulse duration and 250 kHz repetition rate. The interpulse time (τ_{int}) and the number of pulses (N_{pulse}) were tuned by inputting the trigger pulses produced by field programmable gate array (National Inst.; FPGA module 7811R). A series of dots were written in fused silica using the orthogonally polarized femtosecond double pulses varying the delay time (τ_{delay}) between the two pulses. The double pulses were focused via a microscope objective (Nikon; LU Plan Fluor, 100× 0.90 N.A.) at a depth of about 50 μm below the sample surface. The pulse energy of both pulses was about 1.0 μJ and the beam power measured after microscope objective was independent on the orientation of light polarization. After writing, the modified structures were inspected under an optical microscope, a polarized optical microscope (CRi; LC-Polscope), and a confocal Raman spectrometer (Tokyo Instruments; Nanofinder 30).

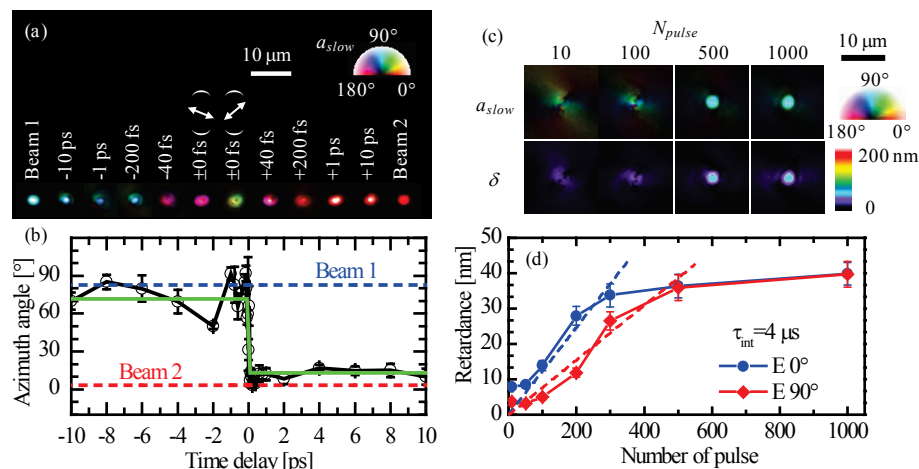


Fig. 1 Characteristic polarized optical micrographs of localized birefringence (a), corresponding plots of the azimuth angle versus delay time between double pulses (b). Azimuth angle in the orthogonally polarized single beams (Beam 1: $E = 0^\circ$ and Beam 2: $E = 90^\circ$) are also shown by blue and red dashed lines, respectively. Green curve in (b) indicates the fit by theoretical functions of the autocorrelation. Polarized optical micrographs of the orientation of a_{slow} and the d induced by the horizontally and vertically polarized light ($E = 0^\circ$ or 90°) versus N_{pulse} (c), corresponding plots of the value of d induced by the horizontally and vertically polarized light ($E = 0^\circ$ or 90°) versus N_{pulse} (d). Blue and red dashed lines are indication of the accumulation speed of the photo-induced d value.

Previously, we reported that the oxygen defects were self-organized in the orthogonal direction to the light polarization [3]. Such nanostructure indicates negative form birefringence with the slow axis (a_{slow}) is perpendicular to the light polarization (E). Fig. 1(a), (b) show time-resolved measurements for a_{slow} using double pulse configuration with the time resolution of 70 ± 40 fs. The a_{slow} approached from the initial value created by the first pulse asymptotically to that of the second pulse. Although small delay time produces birefringence with a larger margin of error caused by the instability of rotation in clockwise and anticlockwise directions, this rapid change can be explained by erasing and rewriting of nanograting structure alternately created by the orthogonally polarized double pulses. Based on the autocorrelation function, the time scale of the modification was estimated to be 70 ± 40 fs, corresponding to the pulse width inside fused silica. These results reveal that the nanostructure can be created by light within the pulse duration of 70 fs. Generally, the oxygen mobility in glass matrix is very slow compared with the optical phenomena. Assuming the temperature of the focal spot reaches 3000°C , the self-diffusion length was calculated by Fick's law. Since the width of the oxygen defect regions in nanostructure was ~ 20 nm, the diffusion time during the motion of oxygen whole length is estimated to be at least $40 \mu\text{s}$.

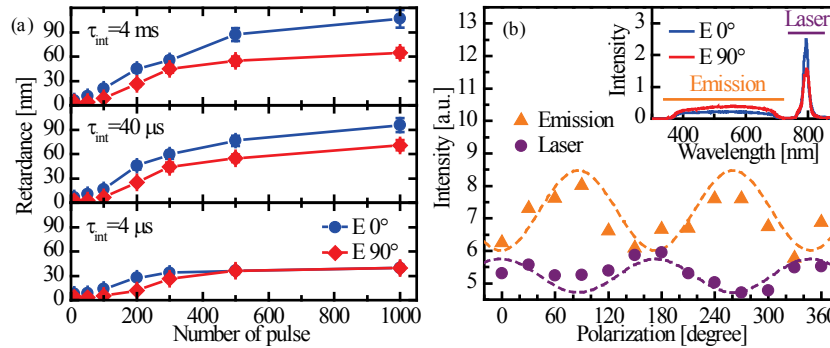


Fig. 2 (a) Difference in the evolution of d between the orthogonally polarized pulses with the different tint of 4 μs , 40 μs , and 4ms. (b) Transmitted peak intensity profiles of the broad white light emission ranging from 340 ~ 720 nm and the incident laser attenuated by the wavelength filter. Sinusoidal fitted curves by sine function are also shown.

In order to reveal the growth mechanism of the form birefringence, we have observed the evolution of retardance varying τ_{int} and N_{pulse} with the same pulse energy of $1.0 \mu\text{J}$. Fig. 1(c), (d) show the evolution of the orientation of a_{slow} and the retardance value (δ) induced by various N_{pulse} at the τ_{int} of 4 μs . Residual birefringence was observed at the irradiated pulses fewer than 100, which was originated from internal stress distribution due to local heating. The apparent form birefringence was observed in the center of the residual birefringence over 100 shots. The growth speed of the photo-induced retardance at the irradiated pulses fewer than 100 at $E = 0^\circ$ or 90° were estimated to 0.14 and 0.06 nm/pulse, respectively. Once the retardance has saturated, simultaneously, a distinct form birefringence has been visible. Interestingly, the difference in the retardance induced by the orthogonally polarized pulses was observed (Fig. 1(d), 2(a)). Such polarization dependence could be interpreted in terms of the difference in the internal stress distribution due to local heating. Indeed, polarization dependent nonlinear absorption and white light emission were also observed (Fig. 2(b)). These results clearly indicate the energy transfer from incident light to plasma emission depends on the polarization (Fig. 2(b)). Since the absorbed laser energy may not depend significantly on the polarization, compared to the white light emission, the observed polarization-dependent anisotropy of light-matter interaction could be interpreted as follows. The same amount of absorbed energy can produce two different types of excitations depending on the laser polarization direction: one is associated with isotropic heating of glass matrix by electrons accelerated via inverse bremsstrahlung and another one is associated with the anisotropic glass heating, cavitation, and defect formation produced by the electron plasma waves excited by the light with the asymmetric relation between pulse front tilt and polarization direction. In our experiments, when the horizontal polarization is aligned along the pulse front tilt, heats less than the orthogonal polarization.

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